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ATMOSPHERIC ACTINOMETRY

AND THE

ACTINIC CONSTITUTION OF THE ATMOSPHERE.

BY

E. DUCLAUX,

*Professor of Physics in the Agronomical Institute, Paris.*

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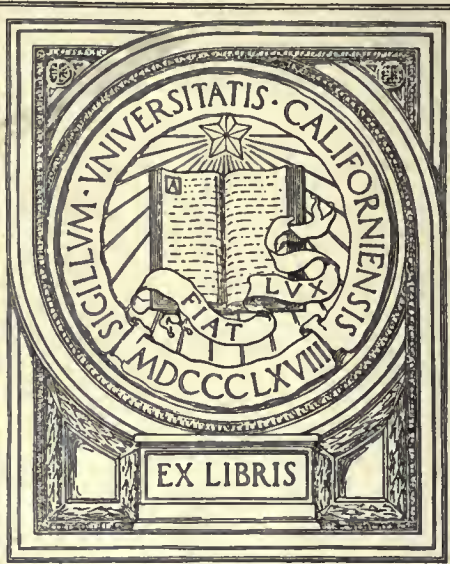
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## ADVERTISEMENT.

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The present memoir is a translation of the treatise entitled "*Sur l'actinométrie atmosphérique et sur la constitution actinique de l'atmosphère*," submitted by Professor Emile Duclaux, in competition for one of the Hodgkins Fund prizes offered by the Smithsonian Institution in a circular dated March 31, 1893. The competition closed December 31, 1894; and on August 9, 1895, the Award Committee, having completed its examination of the 218 papers submitted by contestants, granted honorable mention to Professor Duclaux and recommended his memoir for publication by the Smithsonian Institution.

The Committee was composed of the following members: the Secretary of the Institution S. P. Langley, Chairman, ex-officio; Doctor G. Brown Goode, appointed by the Secretary of the Smithsonian Institution; Assistant Surgeon-General John S. Billings, appointed by the President of the National Academy of Sciences; and Professor M. W. Harrington, appointed by the President of the American Association for the Advancement of Science. The Foreign Advisory Committee, as first constituted, was represented by Monsieur J. Janssen, Professor T. H. Huxley, and Professor von Helmholtz; and after the death of the latter, Doctor W. von Bezold was added.

S. P. LANGLEY,

SECRETARY.

Washington City, May, 1896.



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# ATMOSPHERIC ACTINOMETRY AND THE ACTINIC CONSTITUTION OF THE ATMOSPHERE.

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BY E. DUCLAUX,

*Professor of Physics in the Agronomical Institute, Paris.*

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## ATMOSPHERIC ACTINOMETRY.

The progress made by science leads us more and more to attribute to chemical rays a special action, which is different from and, to a certain extent, independent of that of the calorific and luminous rays. The chemical radiations of the sun, reaching the limits of our atmosphere, become modified while passing through it, according to a law which is peculiar to them ; and, so far as can be seen in so new a subject, their absorption is not the same as that of the calorific or luminous parts of the spectrum.

Photographers, especially those who take landscapes, well know that days which are equally warm or equally luminous do not always give the same results for the same length of exposure, and that there are days when, for some unknown reason, the chemical impression is much slower than on others.

Another argument may be drawn from what often happens in northern lands, where vegetation, which is well known to be specially susceptible to the power of chemical rays, makes much more rapid progress than in temperate regions, notwithstanding the fainter light and the lower temperature.

To what are such differences due ? What law does the chemical absorption of the atmosphere obey, and on what does it depend ? Ought we to attribute it to its normal elements : oxygen, nitrogen, carbonic acid, and water vapor ? Then it should have some general uniformity. Or ought we to see in it, on the contrary, the action of solid or volatile elements, which incessantly reach it from the bare or from the cultivated soil ? Then it should have a local character, leading to a multiplicity of chemical climates. These are very important questions, for

which science has as yet no answer; not that the subject has not already been thoroughly investigated, but because in all the actinometric inquiries proposed so far, sufficient care has not been bestowed upon the separation of chemical action from luminous and calorific effects.

The process which best shows the incorrectness of the methods employed heretofore is that of Messrs. Bunsen and Roscoe, which depends upon a mixture of chlorine and of hydrogen, exposed to the light. The intensity of the chemical action is then estimated by the quantity of hydrochloric acid formed in a given time, or rather by the diminution of volume which necessarily follows. This method has two grave defects. One is that reaction may take place from the effect of heat quite as well as from that of chemical rays, and that consequently it does not separate the two actions which it is important to isolate. The second, much more serious, defect is this, that the reaction is extremely exothermic and continues, when once begun, under the influence of the heat which it develops. There is, therefore, no proportionality between the active cause and the effect it produces. The cause is simply provocative and starts a mechanism, which continues to work independently. It is true that an effort is made to reduce to a minimum the work of this mechanism, by operating only with very small quantities of gas and by multiplying the cooling surfaces, in such a way that the phenomenon constantly requires a new excitation in order to continue. But this is not sufficient to relieve the method of the charge of lacking proportionality between cause and effect, which renders the measurements almost illusory, in spite of the care taken by Messrs. Bunsen and Roscoe to discuss them.

We find the same defects, though perhaps a little less seriously, in the often employed method which depends upon the reduction of ferric oxalate by light. Since the first observation by Dobereiner, H. Draper, Marchand, and G. Lemoine have studied this reaction. As in the preceding case the oxalate, or the equivalent mixture of ferric chloride and oxalic acid, is reduced by the action of heat alone, and although this reduction is slow, it operates as a source of error. Moreover, the liquid is colored and loses its color in proportion as the process continues. Hence the conditions of absorption are modified during the process and this by a phenomenon which is to a certain degree external. Finally, the reaction is still sufficiently exothermic to require that this property should be taken into account. All these defects have been corrected, so far as possible, by M. G. Lemoine, who has for some time been making a careful study of the process, but the method loses thus that neatness and that simplicity which are so desirable.

The ideal would be attained by the discovery of a limpid and transparent liquid which would not change while the reaction went on, becoming the seat of

an easily measurable, chemical phenomenon, which could be the result of no action except that of heat. Can we go any farther in our demands and require that it should not be exothermic in any degree? M. Berthelot does not think so, and believes that the addition of energy resulting from the absorption of calorific, luminous, or chemical radiation would not be sufficient to produce a chemical phenomenon which, while going on, would not give rise to a small amount of heat. I do not very well see why a calorific absorption of solar radiation might not compensate for some heat of combination, and even permit a slightly endothermic reaction to appear. I have endeavored to discover some simple method, which, complying with this programme, could be interpreted without ambiguity, but I have not succeeded. I have been compelled to content myself with an old, well known reaction—the oxidation which weak solutions of oxalic acid undergo upon exposure to light.

These solutions are and remain transparent. The oxalic acid in them is transformed into carbonic acid, which disappears by diffusion, so that the oxidation which it has undergone can be easily ascertained by an acidimetric determination—made before and after its exposure to light. The reaction thus produced is faintly exothermic, to be sure, but as only very weak solutions are taken, for it is well not to exceed 2 or 3 grammes of crystallized oxalic acid per litre, there is no reason why we should be troubled about the error which arises from this fact. Moreover, the liberation of heat, which results from combustion, even if it should be perceptible, would remain without effect, for oxalic acid oxidizes only with extreme slowness under the influence of heat alone.

*Exp.*—10 c. c. of a solution of oxalic acid, titrating 19 c. c. of lime-water per litre, was heated on a water bath to nearly 95°, in a flask of 125 c. c.

After heating 4 hours, the titre is 18.5 c. c. Loss 2.6 per cent.

“ “ 8 “ “ “ 18.0 “ “ 5.2 “

*Exp.*—10 c. c. of another solution, titrating 16.6 c. c. of lime-water per litre, was heated an hour and a half to 115°.

The titre falls only to 16.2 c. c. and 16.1 c. c.

During the days of greatest heat, the temperature hardly exceeds 50° in the shallow vessels, in which the liquid is exposed to the sun. It may, therefore, be assumed that neither the solar heat nor the heat produced by combustion have any perceptible effect upon the transformation of the oxalic acid, which may, on fine days, reach or even exceed 50 per cent of the acid contained in the solution.

As the calorific rays have hardly any power, it would be desirable to eliminate the action of the luminous rays also, but the chemical radiations are so closely intermingled with the latter that it is difficult to separate them. Let us be content,



therefore, for the present, to know that our solution of oxalic acid is peculiarly affected by the action of the luminous and the chemical parts of the spectrum. We shall soon find reasons to believe that it is the chemical part alone which acts. But we have first to investigate the manner by which combustion is produced before we can determine what influences cause it.

#### STUDY OF THE PROCESS.

Oxalic acid, dissolved in water and exposed to light, absorbs oxygen, and changes almost entirely into carbonic acid. There appears also a little formic acid, but in almost infinitesimal quantities. Hence it follows, as we have seen, that we can ascertain the quantity of oxidized acid by a simple acidimetric determination.

#### INFLUENCE OF CONCENTRATION.

In order to study the actinometric process, the first thing to discover is the degree of concentration which gives the largest amount of sensibility. In order to know this I exposed to the sun, under precisely the same conditions, during three fine days from June 4th to June 6th, including about 36 hours of insolation, four liquids, containing, respectively, per litre :

	gram.	gram.	gram.	gram.
	63	31.5	12.6	6.3 of oxalic acid ;
that is,	1	$\frac{1}{2}$	$\frac{1}{3}$	$\frac{1}{6}$ of an equivalent,

per litre. At the end of this time, an acidimetric analysis gave me the quantities of acid which had been burnt, and I computed from this the proportion of acid which had disappeared from each of the vessels.

The figures were the following, counted in milligrammes per litre :

	1 Equiv.	$\frac{1}{2}$ Equiv.	$\frac{1}{3}$ Equiv.	$\frac{1}{6}$ Equiv.
Quantity of acid burnt,	2,500	2,800	4,700	3,300
Proportion,	4 %	9 %	38 %	52 %

To reach the maximum in the absolute quantity of acid consumed we must, therefore, operate with solutions neither too concentrated nor too weak. Solutions which are too concentrated oxidize slowly, and the variation in chemical value is often noted with difficulty. It is, on the other hand, very easy to measure this variation with solutions which are rather weak, because it represents a notable fraction of the primitive value. But, on the other hand, when the liquid has been weakened by the sun, the last portions burn quite slowly. There are, therefore, two dangers to avoid.

After various trials I decided upon a solution whose variation of titre during the most favorable days should not exceed one half of the initial value. This is a

solution containing about  $\frac{1}{2}$  an equivalent or about 3 grammes of oxalic acid per litre. Ten cubic centimetres of this liquid are saturated by about an equal volume of common lime-water, so that the daily variation of the titre amounts to 4 or 5 c. c. of lime-water, a quantity which can be measured down to  $\frac{1}{100}$  by means of a burette. The accuracy which we thus obtain is more than sufficient, as we shall presently see.

#### INFLUENCE OF THE DEPTH OF SOLUTION.

In discussing the question of oxidation, we must consider the part played by the ease with which oxygen penetrates into the depths of the solution. We can easily calculate that the 10 c. c. of oxalic solution, which are used in each one of the experiments, require for complete combustion about 3 c. c. of oxygen, a quantity greatly superior to that which is already dissolved. Whatever the facility may be with which this gas penetrates into a liquid, which is subjected for 8 to 10 hours to insolation in free contact with the air, we may well ask if a solution of oxalic acid oxidizes in the sun in the same way in a vessel of shallow depth, in which its thickness is small, as in a cone-shaped glass or in a round tube. The following experiment furnishes an answer to this question:

*Exp.*—On August 16th, 17th, and 18th, I exposed to the sun 10 c. c. of a  $\frac{1}{20}$  normal solution of oxalic acid, as follows:

- (a) In a cone-shaped glass,
- (b) In an ordinary test tube,
- (c) In a Bohemian glass matrass with flat bottom.

To secure uniformity of temperature, the cylindrical tube *b* was placed upright in the matrass *c*; the exposure continued from 8 o'clock A.M. till 3.30 in the evening. The following proportions of acid were consumed:

	a	b	c
August 16th,	29 %	— %	65 %
“ 17th,	34	14	97
“ 18th,	34	13	84
“ 19th,	31	14	87

Thus, everything else being equal, the proportion of acid consumed is much greater in a vessel with a flat bottom than in a cylindrical tube. The difference is indeed so very striking, that the difficulty with which oxygen penetrates the solution does not suffice to explain it.

A combustion of 13 per cent, produced in 7 hours in the 10 c. c. of liquid, contained in the matrass *b*, has not required more than 0.4 c. c. of oxygen; in other words, about 6 times the normal quantity dissolved in the solution. When



we think of the rapidity with which de-aërated water aërates itself anew, it is hard to believe that it was the oxygen which was wanting, and we are thus led to believe that the chemical action was at fault. If the incident ray does not bring with it an excess of chemical energy, the superficial layers absorb as much as is available, and there is none left for the lower strata, even though all the needful oxygen should be at hand to burn the acid which is present.

The question is of some importance, because it teaches us the quantum of chemical action which may be expected from light in the vicinity of the soil, and consequently, also, the degree of atmospheric absorption. In order to get information on this subject, let us operate with shallow, cylindrical vessels, which are at most a centimetre high at the rim, so that there can be no stagnation of air above the liquid, and that the oxygen always has easy access to the latter. If the actinic influence is deficient in the incident light, we must be able to put in evidence the influence of the surface and the depth of the liquid. For equal depths the combustion will have to be proportional to the surface. For equal surfaces with different depths, combustion, if limited to the superficial layers, should not increase with the volume and the depth of the solution, or at least not increase so rapidly. This is exactly what experience shows.

*Exp.*—Into two cylindrical, very shallow vessels, having the same surface, I poured 10 and 20 c. c. of a half-deci-normal solution of oxalic acid. After a rather dark and somewhat stormy day, I find that 28 per cent. of the acid has been burnt in the vessel that held 10 c. c., and only 23 per cent. in the other. As it held twice as much liquid as the other, the absolute quantities of acid burnt are relatively 28 and 46, while the depths of solution were in the ratio of 1 to 2. Combustion, therefore, increases less quickly than depth. As the latter has not exceeded a centimetre in the vessel in which it was greatest, and as, moreover, the total combustion was very slight, we cannot admit that oxygen was wanting. But the solar rays, deprived of their chemical radiations, which were rendered active by their passage through the superficial strata, reached the lower layers very much weakened, although the luminous transparency of the two liquids was perfect.

There exists then a kind of shifting of the actinic rays during the passage of the light through the first layers which it encounters; and, whether these rays are not abundant or whether the absorption be very efficient and the medium very opaque for them, the weakening process is very rapid. In return, when the surface alone is allowed to vary, while the height of the liquid remains unchanged, the effect of combustion is proportional to the surface, and consequently to the volume.

*Exp.*—I procured two cylindrical vessels with flat bottoms, of Bohemian glass, the bottom surfaces of which were as 1 to 2. I exposed them to the sun, one with 10 c. c., the other with 20 c. c. of one and the same half-deci-normal solution of oxalic acid. The quantities of burnt acid have always been in the proportion of 1 to 2, in a long series of experiments, with an approximation equal to that which the process of analysis should demand.

We shall have to avail ourselves of all these results when we try to ascertain the cause of atmospheric absorption. Let us be content, for the time being, with drawing a practical conclusion from them, namely, that it is desirable always to work with vessels of the same dimensions, and with equal quantities of solution, if we wish to obtain figures that can be compared with each other.

I employ small blown vessels of Bohemian glass, with flat bottoms, such as are found in trade. I choose them of the same dimensions, or nearly so, which can easily be ascertained by fitting together their edges and noting whether they have nearly the same external diameter. It is not necessary to carry accuracy any farther, considering all the inevitable irregularities connected with measurement.

Those which I have used measured about 4.5 centimetres in diameter, and 10 c. c. of liquid had there a thickness of about 6 millimetres. When I was at work in the country, in the Cantal or in the Puy-de-Dome, where the clouds of atmospheric dust are not calcareous, I left them freely exposed to the air upon a small table, so placed as to face the south and to expose them to the sun all day long. The heating which takes place in them is never very great, as the following experiments show; although they were not made in a flat vessel, but in a cone-shaped glass with a foot. The heating is less by 4° or 5° C. when working with a flat vessel.

*Exp.*—The same glass with a foot and containing 10 c. c. of a half-deci-normal solution of oxalic acid was exposed from the 15th to the 27th of August, 1885, daily to the sun. Every day the mean pressure, the maximum temperature of the liquid, and the aspect of the sky were carefully noted. Here follow the proportions of acid burnt on the different days, during which the weather was very fine. The experiments were made at Fau, in the Cantal, at an altitude of about 700 metres.

Date.	Barom. Pressure.	Max. Temper.	Combustion.	Condition of the Sky.
August 15.	712 mm.	36°.8	21 %	Fine. Slight cirrus.
" 16.	712 "	37°.5	29 %	Fine.
" 17.	710 "	37°.5	34 %	"
" 18.	710 "	37°.5	32 %	"
" 19.	705 "	35°.9	32 %	East wind. Clear sky.
" 20.	705 "	35°.2	30 %	" " " "
" 21.	705 "	30°	24 %	Cirro-cumulus. Fresh weather.
" 22.	705 "	25°	24 %	Sky clouded in the morning.
" 23.	705 "	22°	22 %	" " " " Cumulus.
" 24.	710 "	30°	27 %	Sky overcast.
" 25.	710 "	25°	25 %	Stormy weather. Rain at night.
" 26.	705 "	30°	30 %	" " Cumulus.
" 27.	700 "	29°	26 %	" " "

The most active combustions correspond to the highest maximum temperatures, but only because both of them indicate, each in its own way, the presence of a livelier and more active sunlight. When the sky is overcast or shows cumuli, the solar combustion may be more powerful than when there are cirri, even though the maximum temperature should be lower.

In no case, as will be seen, has the temperature of the solution risen to a sufficiently high level to affect the chemical combustion which takes place there. We may, however, if we wish it, secure ourselves against this cause of error by causing the light vessels which contain the solution of oxalic acid, to float on a water-bath. They will then, during the day on which they are exposed to the sun, be heated a few degrees only. This is a method which I have adopted only during the hottest and driest days. The water-bath served as much to restrain evaporation as to prevent heating of the solution.

#### INFLUENCE OF THE AGE OF THE SOLUTION.

We now reach an unexpected fact, namely: that a fresh solution of oxalic acid does not behave like an older solution of the same strength, and appears much more refractory to the action of the sun. It becomes sensitive only very slowly, and it requires even several weeks for that end, when it is kept in diffused light.

*Exp.*—On September 5, 1885, I compared an old solution of oxalic acid, containing  $\frac{1}{40}$  equivalent (1.575 gm.) of this acid per litre, with another liquid, which I prepared at that moment, of the same strength. The common titre of these two solutions amounted to 22.8 c. c. of lime-water for 20 cubic centimetres.

At the close of the day (September 5th), which had been rather foggy, two insulated vessels containing the older liquid titrated together



16.2 c. c. of the same lime-water and had consequently lost 6.6 c. c. of their original strength. Two vessels with the new solution titrated together 21.7 c. c. and had consequently lost not more than 1.1 c. c. The new solution is therefore nearly 6 times less sensitive than the other.

The next day, the weather being fine, the losses amounted to 1.5 c. c. for the new solution and to 8.5 c. c. for the old. This is about the same ratio as on the day before.

On September 12th, after a fine day, four trials gave me the same results, losses of 9.3 c. c. for the old solution, and of 5.9 c. c. for the newly made. The difference in sensitiveness was less marked than six days previously.

On September 25th, twenty days later, the losses became 8.6 c. c. for the first liquid, and 7.7 c. c. for the second. This is not yet equality, which was reached only in the month of October, after a little more than a month.

The fact that two liquids of different ages reach at the end of some time the same degree of sensitiveness, proves that there must be a maximum. We shall, however, soon see that this is not a *maximum maximorum*. However this may be, if the solution of oxalic acid has once reached this maximum, it differs in no way from what it was at first, neither from a chemical nor from a physical point of view; it gives by evaporation the same crystallized acid, and its acidimetric value is unchanged. A molecular activity, however, has been at work, upon which I shall not dwell just here. I will state now only two important facts concerning it: one is that it requires time for its completion, and the other that it betrays itself by easier oxidizability under the influence of solar radiations.

The only phenomenon which in our present state of knowledge may be compared to that which we have just discovered, is the increase of sensitiveness observed in sensitized collodion which has been allowed to rest and to grow old for a few days. This fact is well known to photographers. It may make us think also, by analogy, of the variations in the rotatory power of sugar solutions, some hours after their preparation up to the moment when they become stable. It is admitted that these few hours are necessary to enable the sugar molecules to spread uniformly throughout the solution and to assume the orientation necessary to stable equilibrium. But all these analogies are remote. The phenomenon deserves being investigated by itself, and we have here only to face its practical consequences.

These may be summed up in a few words: that it is desirable to allow the oxalic acid solution to acquire such sensitiveness before using. This is all the easier since

these solutions can become sensitive in a concentrated state and preserve this sensitiveness even after being diluted. One can then provide a mother-liquor, so to speak, which may be made sensitive and which afterwards may be diluted as necessity arises. Ordinarily I used to prepare a normal solution to the amount of several litres, containing 13 grammes per litre, which I kept for some weeks under diffused light, and subsequently diluted, in fractions, to the twentieth or fortieth degree. One litre of this mother-liquid, rendered duly sensitive, may thus serve for 2000 tests. In all the comparative experiments which will be mentioned in this memoir, I have always taken pains to work with identical liquids and such as had the same sensitiveness.

We are now possessed of our actual working process, which amounts to this: To expose to the sun during the day a shallow dish, containing 20 c. c. of a half-deci-normal solution of oxalic acid, which has become sensitive by time, and to measure at the close of the day, by a titration with lime-water, the quantity of acid which has disappeared by oxidation.

Let us now see what results have been obtained by this process.

#### ACTINOMETRIC MEASUREMENTS.

Since the year 1885 I have made several series of actinometric measurements, especially during fine weather and at times when I was sufficiently master of my own time to secure to them the regularity which they require. All these experiments, made at different times and at different places, are not absolutely alike, since the solutions used might have undergone some change. But such variations amount to little from one year to another, and to almost nothing in the course of the same year, as I have been able to determine repeatedly; for every time when I changed the solution, I exposed simultaneously two or more vessels with old and with new material, and I always found that the solar combustion was the same for both, up to that degree of approximation which the measurements demand.

While operating with two or more vessels containing one and the same liquid, it does not always happen that we find the same result for all at the close of the day. There are irregularities in the process, some of which will be explained presently, while the others have until now defied all efforts at explanation, so sudden are they and so exceptional. There is no other remedy for this than to eliminate such out-of-the-way cases, which are always rare, making every day a trial with 3 or 4 vessels and keeping, of the figures thus obtained, only those which are concordant.

It is in this way that the following observations have been made. For each of them a record has been kept of the proportion of oxalic acid burned in 10 c. c. of a



half-deci-normal solution exposed to the sun. Furthermore the state of the sky has been recorded and the principal incidents of the day of insolation.

Special attention has been paid to verifying the solar and antisolar lights<sup>1</sup> which were very frequent during the first years in which these observations were made and which have never been absent since that time. I have described the aspect they assumed in the countries in which I began my observations. The more I study them, the more I consider them as solely due to the presence of aqueous vapor at very great heights in the atmosphere. We shall have to examine, from this point of view, their influence on the phenomena of solar combustion.

The tables which follow are also intended to show the very considerable variation which the quantities of oxalic acid consumed present from day to day. The combustion, which is almost completely absent on cloudy or rainy days, may reach or even exceed 50 per cent of the acid during bright and luminous days. But there are also some very bright days, during which combustion is feeble, and twice it has happened that I was unable to take photographs for want of proper light, being deceived by the apparent brightness of the day on which I was working.

I shall quote my observations very nearly in the order in which I made them, from the moment when I had regulated the process of measurement; and in connection with each one of these sets of observations I shall cite the facts which they have revealed to me and which subsequent observations have only confirmed.

#### OBSERVATIONS OF THE YEAR 1885.

Made at Fau (Cantal). Altitude 800 metres. Country of meadows and of woods. Volcanic soil. (Andesite and basalt of the plateau.)

I have inserted above (page 8) some observations which I made at the end of August with the solution in a conical glass. The combustion is a little less rapid than in the vessels of Bohemian glass which I used in experiments of a later date. Here follow those made in September and October. S. and A. S. represent solar lights in the west and anti-solar lights.

<sup>1</sup> See, on this subject, a note inserted in the *Comptes Rendus de l'Académie des Sciences*, vol. xcix, p. 714.

Date.	Combustion.	Remarks.
September 2	32%	Rain the night before. Clear weather. S. faint.
" 3	0%	Rainy and stormy day. No sun.
" 4	7%	Glimpses of the sun. Barometer rises again.
" 5	13%	Day partly sunny, partly rainy.
" 6	7%	Rain in the morning; a little sun in the evening.
" 7	7%	" " " " " " " " " " " "
" 8	28%	Day in appearance similar to the two preceding.
" 9	7%	Fog and rain in the morning; sun in the evening. S. and A. S. continuous during more than an hour.
" 10	35%	Very fine day. S. and A. S.
" 11	28%	Rainy in the morning. Clearing in the evening. S. and A. S. very bright.
" 12	36%	Fine day, corona around the moon. S. and A. S.
" 13	22%	A fine day. Cirrus. In the evening, at sunset clouds in the west project their shadow in the east upon anti-solar lights. There are besides, in the neighborhood of this light, small cloudlets, the violet-red color of which is exactly the same, except as regards intensity, as that of the anti-solar light. An irregular corona around the moon, fringed and elongated in certain directions by cirrus-streamers to the four points.
" 14	21%	A very fine and very hot day. S. and A. S. very fine.
" 15	27%	A very fine and very hot day. S. and A. S. feeble.
" 16	35%	" day and slight cirrus. No S. and A. S.
" 17	21%	" cirrus and cirro-cumulus. At night storm.
" 18	17%	Quite fine in the morning. Cloudy in the evening. Rain and storm at 6 o'clock P.M.
" 19	17%	A day divided between sun and clouds.
" 20	34%	A fine day. S. and A. S.
" 21	29%	" " Cirrus and cirro-cumulus.
" 22	33%	Fine day without clouds. S. and A. S. fine but short-lived.
" 23	32%	Fine day without clouds. Fine but short-lived S. and A. S.
" 24	29%	Numerous cirri in the evening. Cumulus and storm. Barometer falls.
" 25	1%	Dark and cold day. Lunar halo of 22°.
" 26	13%	Middling day. Some glimpses of the sun.
" 27	0%	Rainy weather in the morning. Dark and cold all day.
" 28	4%	A few glimpses of the sun.
" 29	3%	Fog and rain all day.
" 30	2%	Rain in the morning, in the evening fog. No sun.
October 1	3%	Rain in the morning; very little sun in the evening. S. and A. S. Violet mist in the valley.
" 2	12%	Quite a fine day. In the evening violet lights very perceptible in the neighborhood of <i>Venus</i> , whose brilliancy is very great.
" 3	20%	Sky cloudy all day.
" 4	11%	Fine in the morning; in the evening dark.
" 5	24%	A fine day. Very few clouds. S. and A. S.
" 6	12%	Foggy day.
" 7	1%	Rain all day.
" 8	24%	Fine day, autumn like. Sun rather veiled.
" 9	13%	Rain in the morning; sun in the evening.
" 10	6%	Rain all day. Rare glimpses of the sun.
" 11	13%	Rain in the morning; a little sun in the evening.
" 12	12%	Covered sky; rare glimpses of the sun.
" 13	7%	Sky overcast; cold weather. North wind. Frost at night.
" 14	22%	Fine in the morning, overcast in the evening.
" 15	4%	Dark and rainy day.
" 16	13%	Rainy in the morning; in the evening breaks in the clouds.

Date.	Combustion.	Remarks.
October 17	19 %	Very fine day, from beginning to end.
" 18	18 %	" " In the evening cirrus and halo of 22°. Barometer falls.
" 19	18 %	Dark in the morning, a little sun in the evening. Incessant rain after 3 o'clock P.M.
" 20	5 %	Rainy day. No sun.
" 21	9 %	" "
" 22	4 %	" "
" 23	24 %	Quite a fine day, in spite of east wind which rose very high after 9 o'clock.
" 24	3 %	Rainy day from beginning to end.
" 25	14 %	Partly sun and partly rain. Two currents in the air, one from the south, superior, carrying off cirri ; the other from the north, inferior, with clouds. The latter finally dominates and after having brought up intermittent and slight rains, it gives a cool night.
" 26	2 %	A rainy day.
" 27	15 %	Stormy at night. Day quite fine.
" 28	3 %	Rainy day.
" 29	5 %	" Rare glimpses of light.

These two months of uninterrupted observation prove already that the solar combustion passes through very different values within 24 hours. These changes are sometimes very sudden and exceed especially those of the thermometer, the barometer, and even those of the average brightness of the day. The actinometric effect does not show, therefore, that approximate constancy, which makes it relatively so easy to measure the other effects of solar radiation ; it requires a very close and minute investigation.

While it amounts to little or nothing at all in overcast and rainy weather, it rises very perceptibly during fine, sunny days ; but it seems to be subject to other influences yet beside those which we have mentioned when we spoke of "a fine day," "fine weather," etc. If we find, in fact, that the days from the 20th to the 24th September resemble each other very closely, as far as their external physiognomy is concerned, and are also very much alike in point of actinometry, we have on the other hand the example of October 17th, 18th, and 19th, during which the degree of combustion was the same, and this although the weather had been very fine during the first two days and very indifferent during the last. An instance of the opposite nature is offered to us by the 6th, 7th, and 8th of September, which differed very much in their actinometric aspect, whilst they resembled each other so far as their external physiognomy was concerned.

It would be interesting to find out under what influences these variations are produced. In the meantime, until we reach that point, let us notice that the combustion on the finest days in October does not amount to as much as that obtained on the finest days in September, and that the latter again do not equal the fine



days in August, mentioned on page 8, if we bear in mind that the experiments mentioned on that page were made in a cone-shaped glass, capable of holding the bulb of a thermometer and not, like those made in September, with shallow vessels in which the figures would have been much higher.

One might be tempted to see here the effect of the lessened length of days. But, in order to avoid this influence, the length of exposure has been everywhere precisely the same: from 8.30 A.M. to 4.30 P.M. There is, therefore, an influence due to the seasons, which we must also endeavor to trace back to its true cause. For this purpose we can only collect the greatest possible amount of evidence.

#### RESULTS OF OBSERVATIONS MADE IN 1886 AND 1887.

I made for this end several series of experiments in 1886 and 1887, at Paris, in the Cantal, and at Orcines, at the foot of the Puy-de-Dome. Unfortunately I cannot report them here in detail, having mislaid the papers which contained the record. I can only indicate the general results which I have retained in my memory, because they served as a starting-point for new investigations. In the first place I again found evidence of the almost perfect independence between the degree of solar combustion of oxalic acid, and the occurrence of solar and anti-solar lights. If there are any examples of coincidence between an active combustion and the presence of such lights, it is because these lights appear only in fine weather. But there are also other cases in which combustion is very rapid and when those lights are altogether missing. If they play any part at all, it seems to be one quite secondary, and this view agrees very fairly with the hypothesis that the phenomenon is due to the presence of aqueous vapors in the upper regions of the atmosphere. It is, of course, well known that liquid water or water in the form of steam influences the activity of actinic combustion very little.

Another very important result is this: that the maxima of the figures of combustion during the finest days are higher in spring than in summer. The difference did not strike me as quite so marked as between summer and autumn. As to the maximum in spring I have always found it very clearly marked during observations carried on for four years, and an example of this will be found, unfortunately too limited in its nature, when I shall speak of my experiments of 1888.

The maximum in spring appears alike in Paris and in the country. But I have also found that solar combustion was less intense in Paris than in the Cantal or in the Puy-de-Dome; this difference appears not only in the high figures connected with the oxalic acid, but I have found it also in a long series of experiments, which I had undertaken in order to study the transformations which

several organic substances undergo in the solar light, and which offer a greater resistance than oxalic acid. These I had been compelled to leave in the light for weeks and for months before the attack was complete. They thus summed up the influence to which they had been subjected during the length of the exposure. Now this phenomenon required, generally, for its termination, much more time, sometimes three or four times more, in Paris than in the country.

Among the facts of this kind I can only quote one which I find I had by chance inserted in a work intended for the examination of another question. A deci-normal solution of tartaric acid which was every day exposed from 10 A.M. till 2 P.M. to the sun in Paris, had lost by combustion in seven months and a half only 10 per cent of its acid, while in the Cantal an identical solution had lost in two months 47 per cent. This involves a combustion about fifteen times more rapid, and although the length of exposure was a little greater every day in the Cantal than in Paris, and although the quantum of solar combustion increases more rapidly than the length of exposure, this is not enough to make up for the difference. In another case involving the combustion of glucose in an alkaline liquid, I found that to take two years in Paris which had required only three months in the Cantal.

Finally, this experiment teaches us also that average years do not resemble each other and that, if there are some which are rich in chemical radiations, there are poor ones also. These differences between one year and another, from this point of view, appeared to me more marked than in any other respect. We have shown above the most striking inequalities between consecutive days of the same season. They recur, less markedly, for consecutive years.

These statements which, I repeat, I regret not being able to support by figures, suggest a number of problems for which I have begun to seek a solution.

In the first place, the fact that active combustion is stronger on fine days in spring than in summer and in autumn, shows that there must be another cause of action than the influence of temperature, or the height of the sun above the horizon. We are naturally led to think of the influence of the volatile organic products which vegetation scatters in the air during summer, and which, if they are capable of being oxidized, absorb and utilize for their own benefit the chemical radiations of the light which passes through the atmosphere, preventing them from reaching the soil. We are confirmed in this view by what has been said before (page 6) concerning the relative poverty of solar light, at the time when it reaches us, in radiations able to oxidize oxalic acid.

In the second place, the difference between the sum total of the annual radiations at Paris and at the Cantal, or on the high table-lands of the Puy-de-Dome,



leads us to ask if the question of altitude may not perhaps be of importance. Two identical vessels, containing the same solution and exposed during the same time at different heights in the atmosphere—will they or will they not undergo the same degree of oxidation?

#### INFLUENCE OF ALTITUDE.

I begin with the last question, because the documents which helped me to solve it were lost, together with those which gave the results of the experiments already mentioned, and I must therefore be very brief in my treatment.

In order to solve this problem I installed myself at the foot of the Puy-de-Dome in the little village of Orcines, and I made a number of experiments simultaneously in the garden of the house in which we lived, and on the terrace of the observatory on the Puy-de-Dome, where M. Humandon kindly undertook to expose and to remove again at certain fixed hours the vessels containing the oxalic acid which had been rendered sensitive. The two stations are distant from each other 4 kilometres in a direct line. The vertical difference amounts to about 400 metres. The incline, therefore, between the two stations does not count for much, and they cannot be considered as being upon the same vertical line. Experiments made on the top and at the foot of the Eiffel tower would have been more satisfactory in this respect. But at the Eiffel tower I should have had to apprehend encountering difficulties of another kind, especially the want of homogeneousness between the layers of the atmosphere. For the lower ones which had swept populous parts of the city could, in that amount, no longer be considered equivalent to the upper parts. What tempted me to choose the station of the Puy-de-Dome was exactly this homogeneousness of the whole region so far as its vegetation was concerned. The Puy-de-Dome is surrounded to a great distance by a dry, almost deserted country, covered with woods and largely with heather, while some portions are absolutely bare in places where pozzolanes crop out of the soil or in those immense overflows of lava of recent date, which are called "cheires," and which defy any attempt at cultivation. One of these "cheires" cropped out close to the house in which I dwelt, and I imagine that, on the whole, there was no reason why the air on the top and that of the table-land from which the mountain rises, should be heterogeneous. In spite of these favorable circumstances I did not find that the combustion at the observatory was very different from that at Orcines. It exceeded the latter a little, on an average, but with exceptional results in one or the other direction, so as to prevent any positive conclusion. I remember that my estimates showed an increase of altitude accompanying an increase of actinic intensity, but

that they were not such as to prove it. This conclusion diminishes my regret at having mislaid the data.

INFLUENCE OF OXIDIZABLE SUBSTANCES SUSPENDED IN THE AIR.

With this second question I have been more successful, since comparative experiments can here be made with far more precision than elsewhere. All that is necessary is to expose in one and the same place two vessels containing the same oxalic solution; one being made to float upon water contained in a deep crystalizing pan in such a way that a layer of stagnant aqueous vapor may be kept above the surface of the liquid which it contains. The other vessel floats in like manner, and under the same conditions, on the surface of some turpentine or of any other essential oil. It is always found that combustion is far less advanced in the second than in the first vessel. As I said before, I lost the relative figures of the results obtained by the experiments made in 1886 and 1887. But Mr. Elfving, professor at the University of Helsingfors (Finland), to whom I had mentioned the results thus obtained, began once more to experiment with essence of turpentine, and I will here quote the results as he reported them to me in a letter which I have fortunately preserved:

"I have repeated and confirmed your experiments on August 30, 1888, from 8 A.M. to 4 P.M. with a clear sky. There were 53 per cent of the oxalic acid burnt above the water, and 39 per cent above a bath of essence of turpentine. The next day, which remained clear from 9 A.M. till noon, the figures were 47 per cent and 20 per cent for the same length of exposure. It is certain, therefore, that the presence of oxidizable substances in the air possibly diminishes the consuming power of the sun."

Mr. Elfving has confirmed this conclusion by the following experiment, which I have, in my turn, repeated and confirmed. It consists in sifting, so to speak, the rays of the sun through a solution of sulphate of quinine, which absorbs a part of the chemical radiations before they can react on the oxalic solution. Another sifting apparatus, of the same thickness, but consisting of pure water, furnishes a standard of comparison. The latter, rigorously, might be neglected, for the quantity of watery vapor or of liquid or solid water which the rays of the sun have traversed before reaching the level of the soil, exceeds by far the thickness of the supplementary screen of liquid; the absorption due to water, is, moreover, very feeble. In my experiments I suppressed this complication. Mr. Elfving used two glass bell jars with double walls, of which one contained water, the other a solution of sulphate of quinine. He wrote to me on June 17th:



"The light which has gone through a layer of water is five times more active than that which has traversed a solution of sulphate of quinine of quite the same depth. I shall continue my observations at the time of the solstice."

And on July 9th:

"I have again made an experiment with sulphate of quinine. On June 27th, while there were consumed in the open air during the whole day 87 per cent of the total amount of oxalic acid, and 78 per cent under a bell jar filled with water, the decomposition amounted only to 20 per cent under a precisely similar bell filled with a solution of sulphate of quinine."

Analogous results are obtained by comparing the effect of sifting solar rays through a solution of potassium bichromate, which by preference allows those radiations to pass which are least refrangible, with that of transmission through a solution of sulphate of copper which allows the most refrangible radiations to pass. All this proves that it is mainly the chemical radiations which are of importance, and that when these radiations are employed in oxidation, or more generally in the transformation of organic or even mineral substances in the air, they reach the surface of the soil much weakened.

Here is, therefore, a local cause of variations in the actinometric degree; a local and incidental cause, considering that it may be summed up thus: There may exist actinic clouds, clouds scarcely visible to the naked eye and not accessible to our senses, but the effect of which, at least as far as it can be measured by solutions of oxalic acid, exceeds by far, in intensity, that of the variations in brilliancy and obscurity produced by ordinary clouds. These clouds come and go, are no longer to-day where they were the day before; they dissolve, for they are, like other clouds, no sooner formed than they become subject to ceaseless causes of destruction. This explains very fully why the actinometric degree should vary so greatly from day to day and from one year to another. It may also be that we find here our explanation of the greater actinic power which spring has, in other words, that season during which the atmosphere is certainly poorest in organic substances.

Upon reaching this point we see new vistas open before us. It is well known that the turning green of the foliage and the production of chlorophyll may take place when the intensity of light is very feeble, as for instance in the back of a room lighted by one window, but that, under such circumstances, the chlorophyll does not begin to act and is not decomposed by carbonic acid. It requires a much stronger luminous intensity for the process of assimilation to begin. This phe-

nomenon increases with intensity of light up to a certain maximum, differing in different plants; beyond this it decreases. Starting from this point, it is naturally suggested that those actinic clouds which we have just discovered, cannot be without influence on vegetation, since they modify so largely, although often invisibly, the strength of the chemical radiations, which is nearly, if not absolutely the only active element in the complex whole, which until now has been studied under the name of *Luminous Intensity*. And if again the plants themselves disperse into the air the materials which absorb the solar, chemical radiations, how can we avoid thinking that possibly the production of these odoriferous and oxidizable effluvia may be for the plant a means of protection?

To elucidate this subject fully, would require experiments which I have not the time to make. I have been satisfied with examining it under various aspects. Odorous and essential oils are not alone able to arrest the passage of chemical radiations. The surfaces of plants are, as is well known, frequently covered with a fatty or waxy layer, which is highly oxidizable. There are, besides, at all times fatty substances in the air, as is proved by the greasy feel of old dust deposited upon our furniture. What effect can these fatty substances exert upon the combustion of oxalic acid in the sunlight? If our ideas are correct, a slight layer of fatty matter should protect that acid against solar light.

#### INFLUENCE OF FATTY SUBSTANCES.

The presence of fatty matter on the surface of our test solution brings up a slight experimental difficulty. It is this, that solar oxidation of a fatty substance is always accompanied by a production of acid which raises the titre of the oxalic solution at the same time that the solar combustion lessens it. We must, therefore, either use a very small quantity of fatty matter so that it may barely form an imperceptible veil to cover the liquid, or, what is better still, we must spread it out in a transparent layer over a surface of glass interposed in the path of the luminous rays. Here are some experiments made in connection with this subject:

*Exp.*—On June 27, 1885, I exposed to the sun during six hours seven vessels of the same dimensions, containing each 10 c. c. of oxalic acid in half-decimal solution. Two of these vessels, Nos. 1 and 2, had their walls clean. Vessel No. 3 had been rubbed with a weak solution of butter in sulphide of carbon, which left upon the sides, hardly tarnishing them, a greasy layer. Moreover, by virtue of a well-known phenomenon of superficial tension, the walls of the vessel have allowed an invisible layer of fatty matter to spread on the surface of the liquid. In order to separate

this action as far as possible from that of the roughness of the walls, a fourth vessel is brought up to the same degree of opacity as No. 3, by rubbing it externally with chalk diffused in water. Finally, to increase the quantity of fatty matter present in the liquid, and also, in order to see the effect which a little opacity of the liquid may there produce, new vessels, Nos. 1<sup>1</sup>, 2<sup>1</sup>, and 3<sup>1</sup>, have been prepared like the vessels Nos. 1, 2, and 3, simply adding to each two drops, in other words 5 milligrammes, of fatty matter.

These were the results:

		Combustion.
Vessel No. 1, clean sides.....		33 %
" " 2, " " .....		33 %
" " 3, dull sides, greasy surface.....		29 %
" " 4, " " (chalk).....		32 %
" " 1 <sup>1</sup> , like 1, plus 2 drops of milk.....		16 %
" " 2 <sup>1</sup> , " 2, " " " .....		17 %
" " 3 <sup>1</sup> , " 3, " " " .....		17 %

Other experiments, made the following years, and the detailed reports of which have been lost, confirm these first results, and show that the fatty matter contained in a liquid or spread as an invisible layer over the surface, like that which covers the walls of a bell jar placed over the vessel containing oxalic acid, diminishes the actinic effect of the solar rays.

Finally, it is the same with many substances, more or less easily oxidizable, which also exert a protecting, or at least a retarding, effect upon the influence of the chemical radiations. Such is, for instance, alcohol.

*Exp.*—On the 26th June, 1885, two vessels with 10 c. c. of an oxalic acid solution, containing  $\frac{1}{15}$  equivalent per litre, gave me a combustion of 37 per cent, the same for both. Two other vessels, exactly alike, which have received an addition of 2.5 c. c. alcohol of 90 per cent, gave me only a combustion of 21 per cent.

*Exp.*—On September 14, 1888, two vessels with a solution of  $\frac{1}{20}$  equivalent of oxalic acid, gave me identical combustions, rising as high as 10 per cent. They amounted only to 4 per cent in two like vessels, to which a few drops of oil of oranges had been added, so that the essential oil and the alcohol have acted similarly.

I have made numerous experiments with divers substances which were oxidizing or oxidizable, the details of which have been lost. In a general way I have found that the former increase the combustion of oxalic acid, while the others retard it.



But this rule is not always confirmed, on account of the intervention of what I have called in another paper phenomena of entanglement. These operate so that one oxidizable body may involve another in the same decomposition which it undergoes itself. But here we begin to stray away from the subject of this paper, which is the actinic study of the atmosphere. I am content, therefore, to draw from the great sum total of the results I have obtained the following conclusions: The nature and the proportion of the oxidizable elements which living nature scatters through the air, betray themselves in the solar combustion of oxalic acid, which is the more feeble on the surface of the solution the more the radiations have met with unstable elements to oxidize during their passage. The organic substances of the atmosphere are therefore a protection against too intense an action of the chemical rays at the surface of the soil, and the effect which they produce is not only measurable, but sometimes very great. In other words, we do not know what the chemical power of solar light may be at its entrance into the atmosphere, but on a level with the soil it is so impoverished that a thin layer of turpentine vapor, of sulphate of quinine, or of any oxidizable substance, suffices to destroy it almost completely.

This conclusion has, however, another side to be considered, which is, that the atmosphere must at every moment be the seat of combustions, such that, on the whole, all luminous radiations are utilized. I shall not insist here on the power and the importance of the phenomena of oxidation which take place in the atmosphere and at the level of the soil, nor upon the general effect which they have on sanitation over which they preside. I have published several memoirs on that subject,<sup>1</sup> to which I must be content to refer. I have there called attention to the power of the solar rays on microbes, first weakening and then killing them, a power which was first indicated, but incompletely proven, by Messrs. Downes and Blunt.<sup>2</sup> I have, moreover, studied the influence of the conditions of the medium on the resistance of germs. All that has been done since, has only confirmed the importance which I attached to light and to the chemical portion of the solar spectrum as principal agents in the hygiene of the globe.

#### INFLUENCE OF INCREASE OF LATITUDE.

This first problem, that of the possible influence exerted by oxidizable substances while in suspension in the atmosphere, having been sufficiently discussed in

<sup>1</sup> *Annales de Chimie et de Physique*, 6th S., vol. v., May, 1885, and *Comptes Rendus*, vol. c. and ci. *Annales de l'Institut Pasteur*, vol. i., p. 88.

<sup>2</sup> The conclusions of these scholars had been opposed by Tyndall and by Jamieson, so that when I took up the question anew, it had not yet met with a solution. It has found one to-day.

the statements just made, I found myself face to face, as Professor of Meteorology, with the following question :

It is an acknowledged fact, that the activity of the vegetative process in the northern parts of Europe is very great. The interval between sowing and harvesting, for spring wheat, lasts on an average 145 days in Alsace. According to M. Tisserand it amounts to only 133 days at Halsn , in  $59^{\circ} 30'$  N. latitude ; and it is only 114 days at Skibbotten, in  $69^{\circ} 30'$  N. latitude. It decreases therefore as we approach the pole ; notwithstanding that the average temperature of the period of vegetation diminishes likewise with the increase of latitude.

This decrease in the number of days needed for vegetation, as we draw nearer the north pole, seems to be a general law. According to Arnell, barley requires 117 days to grow in Southern Sweden, 92 in Middle Sweden, and 89 in Lapland. It is true that these variations depend in part at least on the power which the plant has to adapt itself to external conditions, for if sown in our country the Norwegian grain grows more rapidly than ours, while our own native grain, carried to Norway, lags behind the acclimated variety. But this is not sufficient to explain all, and we must in the end always return, as a final analysis, to the influence of climate.

We may go even a little farther in our induction. According to Griesbach the increased rapidity in the development of plants cultivated at the extreme north does not affect the whole evolution of the plant, but only the period between germination and blooming. It applies, therefore, only to the green organs of the plant, and thus starts once more the question of light, which actually appears to be of greater importance than that of temperature. In fine, to return to the subject of our Memoir, the actinic influence of the solar rays seems to increase with the latitude.

To what is this increase due ? This question is still open and to it I have tried to find an answer. The first point to determine was whether the solution of oxalic acid also showed such an increase of actinic effect ?

It was on this account that I asked Mr. Elfving to assist me, whose interesting experiment I have mentioned above (page 17). I sent to him at Helsingfors an oxalic solution, and ten vessels exactly alike, such as I had used myself in France, in order to make sure that at least, and as far as possible, those experimental conditions which we could control should be as identical as they could be made.

Unfortunately there were other conditions which were entirely beyond our control. The ideal would have been attained with a series of days equally fine, occurring simultaneously in France and in Finland, and permitting us to make our observations under precisely the same circumstances. But there are obvious reasons why the weather could hardly ever be the same in France and in the Gulf of



Bothnia. When that vast mass of air, in relative repose, which I have named The Isle of Calms, rests over our part of the world and gives us fine weather, the equatorial current, which turns it northward, is over Sweden and Norway, to which it brings overcast skies and rains; when, on the other hand, the Isle of Calms rests over the north of Europe, we are in France subject to stormy disturbances which come to us through the Mediterranean or the Gulf of Gascony, or we are subject again to the return current, which, after having rounded the "Island," comes back to us in the shape of cold east and northeast winds. To find favorable coincidences in this grand atmospheric dance, we should need months of continued observations, which neither Mr. Elfving nor myself were in a condition to undertake.

In this difficulty we availed ourselves of the meaning of the word "fine day," as I have shown above (page 13), which is so uncertain as to its actinometric definition that every effort to make it absolute as to perfect equality of experimental conditions becomes rather illusory. We could be content, more modestly, with a first approximation; it was enough to compare the actinometric combustion of the finest days in the Gulf of Finland and in France, at the same time of the year.

Nor is this all. The length of the day is greater at the North than at the South during the period of vegetation, and the length of the insolation has, we all know, a direct influence on the relative quantum of combustion. Hence I requested Mr. Elfving to make every day two sets of experiments, one with vessels exposed to the sun from 8 A.M. to 4 P.M., like those which I was using in France, and the other with vessels left out from 8 A.M. till the setting of the sun.

Mr. Elfving made at Helsingfors between August 27th and September 4th, 1887, five series of experiments, which I cannot compare with those which I was making at the same time at the foot of the Puy-de-Dome, and the records of which have been lost. But I am fortunately able to compare them with those which I had begged M. Ch. Mascart to make at the same time near the seashore in the Channel. These may perhaps be better fitted for comparison with those made by Mr. Elfving, as both were made at maritime stations. All that I noted when I received them was that they gave much higher figures than those which I obtained at the same time on the bare table-land which carries the Puy-de-Dome.

In the first place, here is a table of the observations made in France; it is formed in the same way as those which have already been given in this Memoir.



ST. PIERRE LE PORT, 1887.

Date.	Combustion.	Remarks.
August 15	44 %	Clear weather till 10 o'clock P.M. ; later cloudy.
" 16	38 %	Rain till 2 o'clock ; later overcast.
" 17	31 %	Very clear from 11 till 3 ; afterwards cloudy.
" 18	23 %	Three-fourths cloudy till 10 A.M. Clear from 10 till 3 P.M.
" 19	33 %	Half overcast in the morning ; then quite clear.
" 20	21 %	Half overcast all day long ; a slight fog.
" 21	28 %	Slightly overcast in the morning ; then clear.
" 22	30 %	Slightly overcast in the morning ; then clear.
" 23	36 %	Fine weather.
" 24	29 %	" "
" 25	42 %	Warm. Very close. Clear weather.
" 26	32 %	Covered in the morning and evening. Clear from 12 till 3 P.M.
" 27	23 %	Overcast. Rain from 10 till 11 A.M.
" 28	24 %	Unceasing rain.

Here again the solar combustion increases with the fine weather and diminishes when the sky is overcast or rain falls. Although the weather was on an average less fine than during the corresponding series of experiments cited before, the latter gives, on the average, higher results, a fact which confirms what we have already said concerning the actinometric differences of different years at the same epoch.

Here are now the experiments made by Mr. Elfving at the same time in 1887 :

Helsingfors, latitude 60° 10'. Length of day, 14 hours. Height of the sun above the horizon at noon, about 38°.

Date.	Solar Combustion.	
	From 8 A.M. to 4 P.M.	All day long.
August 27	42 %	55 %
" 28	50 %	65 %
" 29	53 %	61 %
September 2	74 %	87 %
" 4	77 %	89 %

"The difference between the first three days and the two others is quite great ; it arises, no doubt from the fact that the atmosphere had been purified by heavy rains on August 30th and September 1st and 3d. In March, I had already observed this effect of rain."  
(M. ELFVING.)

The figures in the first column are on an average higher than those which correspond to them in the preceding report, and this superiority must be all the more

striking, as by a mistake in our agreement the exposure to the sun lasted an hour longer in France than in Finland. The latter ought, therefore, to be somewhat increased, in order to make the comparison more just. We shall presently return to the results marked in the last column. Mr. Elfving had worked only during five days; the comparative experiments were therefore not numerous enough, and by a common understanding another beginning was made in 1888.

Here is the report of the experiments which I made in France, in the garden of the Agronomie Institute, during the months of May and June, 1888. My official duties prevented me from making them in an unbroken series, and, moreover, I had to leave out three observations during which a fierce wind suddenly sprang up and covered my vessels with a layer of dust.

## PARIS. EXPOSURE FROM 8 A.M. TO 5 P.M.

Date.		Combustion.	Remarks.
May	12	46 %	A fine day. Fresh north wind.
"	13	29 %	Very fine day; rather warmer than the day before.
"	14	50 %	Cirrus in the morning. Very fine day.
"	15	23 %	Sky overcast. Barometer falls.
"	17	52 %	" " South wind. Cirrus.
"	18	23 %	" " " "
"	20	27 %	Quite a fine day. N. wind. Cirrus and alto-cumulus.
"	21	35 %	Sky overcast. Lighter in the evening. Fresh east winds.
"	26	43 %	g. g. clouds; fine at night. Fresh north wind.
"	27	30 %	Quite a fine day. g. g. Cirrus. Sudden storm.
June	1	33 %	Fine day. No wind.
"	2	55 %	Warm and stormy day.
	3	39 %	Very warm day. South wind.
	5	42 %	Warm and stormy day. Sky overcast.
	12	64 %	Fine day. A little air.

The correspondence between the degree of combustion and the state of the atmosphere is less striking in these observations, which were made in Paris, than in those made in the country, which is less surprising when we bear in mind the incessant heterogeneity and variability of the air in a large city. The influence of the spring season, however, to which reference was made before (page 14) is shown in the relative magnitude of the figures of combustion. The figure 64 %, dated on June 12th, is very exceptional.

Here follow next the results obtained almost simultaneously by Mr. Elfving at Helsingfors :

## HELSINGFORS. EXPERIMENTS MADE IN 1888.

Date.	Solar Combustion.	
	From 8 A.M. to 4 P.M.	All day long.
May 19	50 %	— %
“ 21	47 %	58 %
“ 22	56 %	76 %
x “ 23	53 %	65 %
x “ 24	37 %	55 %
x “ 27	44 %	— %
“ 30	46 %	72 %
“ 31	51 %	72 %
June 4	48 %	63 %
x “ 7	48 %	70 %
“ 8	— %	74 %
“ 9	56 %	79 %
“ 10	57 %	77 %
“ 11	54 %	80 %

On the days marked with a cross (x), the sky was more or less overcast at Helsingfors. All the figures in the second column ought to be raised slightly, in order to make them fit to be compared with those in the preceding table, which correspond to an additional hour of exposure. It will be seen, however, that they are on an average higher, although none of them reach the exceptional figure of the 12th of June at Paris.

The conclusion is the same as that derived from the experiments of 1887. In order to add to its weight we recommenced another series in August and September. This time I installed myself on the Mont Dore, at a height of about 1100 metres, in a house some distance from that little village.

## MONT DORE, 1888.

Date.	Combustion.	Remarks.
August 9	26 %	Cirrus in the morning, which increased towards evening.
“ 10	19 %	Fine day.
“ 11	18 %	“ Sky slightly covered.
“ 12	19 %	“
“ 13	18 %	Cumulo-cirrus in the morning. Fine afternoon.
“ 14	27 %	Fine day. Sky a little cloudy in the evening. South wind.
“ 16	27 %	Very fine day. Atmosphere limpid. Cirrus in the evening.
“ 17	—	Rain all day. No exposure.
“ 18	—	Sky overcast, and rain.
“ 19	22 %	Fine day. Cumulo-cirrus and cirrus.
“ 20	15 %	Cirrus all day, especially in the evening. Barometer not falling.
“ 31	15 %	Large white cumuli.



Date.	Combustion.	Remarks.
September 1	15 %	Same weather as day before.
" 2	10 %	Middling day.
" 3	24 %	Quite a fine day. Many cirri.
" 4	12 %	Middling day.
" 5	11 %	"
" 6	12 %	Quite a fine day. Some cumuli early.
" 7	11 %	Middling day.
" 10	11 %	Quite a fine day with a few clouds.
" 11	7 %	" " " "
" 12	18 %	Fine day, hot sun, a few cumuli.
" 13	25 %	Fine in the morning, middling later.
" 14	15 %	Fine day, very warm.
" 15	9 %	Very fine day, as yesterday.
" 16	4 %	Middling day. Warm and heavy.
" 17	29 %	Day divided between sun and clouds.
" 18	—	Dark day.
" 19	30 %	Superb day.
" 20	10 %	Sky fine early ; covered in the evening.
" 21	17 %	Day rather finer than day before.
" 22	13 %	Quite fine in the morning. Cloudy at night.
" 26	15 %	Cumulus concealing about $\frac{1}{3}$ of sky.
" 27	25 %	Rather better than the day before.
" 28	49 %	Rather a dull day, but no clouds.

Between the 20th and 30th August there followed a long period of rain and overcast sky.

What strikes us in reading these figures is their smallness even on fine days. They are the smallest I have ever had to record in August and September, on an average, and this although the latter month was rather fine at Mont Dore during 1888 ; there is also to be noticed a great lack of agreement between the apparent character of the day and its actinometric character. Thus the very fine day of September 15th gave only a combustion of 9 per cent, when the slightly veiled day of September 28th gave a combustion of 49 per cent. This is a new confirmation of what has been stated before.

I partly attribute the very great want of agreement noticed at Mont Dore to the fact that this station is surrounded on all sides by pine woods which diffuse through the air a large quantity of terebinthine exhalations, so that the odor becomes striking. This explanation also agrees with the notions which I have suggested before. Nevertheless I admit that it would require very special comparative experiments to establish it firmly, and to draw from it the proper signification. We must be content, for the present, to remark that if our explanation is correct, it will also account, as a whole, for the want of agreement already mentioned. If the exhalations of essential oils are really able to arrest actinic radiations, the effect of what we call a fine day will be very variable according as it will succeed a period of rains which may have washed the atmosphere, as in the

observations made by Mr. Elfving, or as it may come to us after a warm day or a period of great heat, which may have increased the invisible cloud of terebinthine vapors or other odorous essences. But, I repeat, all these points must be investigated directly, and this preliminary study, although it has continued for many years, has no other claim than that of suggesting new subjects for the study of the atmosphere.

Let us now return to the comparison of the effects which equal periods of exposure have in France and in Finland. The following are the results obtained by Mr. Elfving at Helsingfors, during the same period of the same year :

HELSINGFORS, 1888.

Date.	Solar Combustion.		Remarks.
	From 8 A.M. to 4 P.M.	All day long.	
August 22	56 %	66 %	Clear sky.
" 23	51 %	60 %	Almost clear
" 26	35 %	45 %	Cloudy.
" 27	56 %	75 %	Clear.
" 28	50 %	68 %	Half-overcast.
" 29	55 %	71 %	Very clear sky from 9 A.M.
" 30	53 %	70 %	Very clear sky from 9 A.M. to 12.
" 31	39 %	—	" " " " "
September 2	49 %	59 %	Very clear sky.
" 3	49 %	67 %	" " " "
" 6	54 %	—	Almost clear.
" 8	49 %	—	Clear early, overcast afterwards.
" 9	52 %	62 %	Very clear sky.
" 10	56 %	÷	" " "
" 11	59 %	—	" " "
" 14	51 %	—	Clear sky.
" 15	46 %	—	"
" 16	51 %	—	"
" 17	51 %	—	"
" 18	42 %	—	Clouds.

The regularity is here greater than in France, and what is especially remarkable is the close resemblance in an actinometric sense of the days which are marked as "similar," in the column of "Remarks" (Aug. 29th and 30th, Sept. 2d and 3d, 14th, 15th, 16th, and 17th). But what is perhaps most striking in this table, when compared with that on page 26 is that the figures of solar combustion are notably higher than they were in France at Mont Dore, at the same time of the year. Still, Finland is very rich in resinous woods, and even if the station were less surrounded by them than at Mont Dore, the altitude is lower, which to some extent

makes partial compensation. Besides, the higher figures obtained at Helsingfors, persist at the three positions of the French station : at Paris, on the coast of the Channel, and on the mountain of Puy-de-Dome.

There can, therefore, remain no possible doubt on this point: the actinic intensity of light in northern countries, close to the soil, is greater than in our temperate zone at the same hours of the day. It would no doubt be found still weaker if we approached more nearly to the equator. This conclusion was altogether unforeseen.

The fact once established, explanation becomes necessary. This greater activity of combustion which the air has in northern regions, might be ascribed to ozone, rendered more abundant there by the discharges which constitute the aurora borealis, and more active by the action of light. I have begun to study this subject, but my experiments, interrupted by winter and my return to Paris, are not yet completed, and I shall not be able to take them up again till next spring. I believe, however, that I may already say that ozone can have but a very secondary influence on the phenomenon, and that if light is more active within the same length of time at the north than in France, the reason is that it has lost fewer of its chemical radiations in passing through the atmosphere, because the latter is poorer in oxidizable substances. I know, of course, that there are in the north those pine forests, of which I have spoken before, and that perhaps, if Helsingfors were built in the heart of the woods, instead of being a large city on the sea-coast, the points of difference would be somewhat less. But there would always remain the fact that the quantities of vapor diffused through the air increase with the temperature, and that, for one and the same aspect of the fauna and the flora of the earth, the equatorial atmosphere will always be more heavily loaded than that of the temperate zones, which in its turn will again be more so than that of boreal regions.

Whatever finally the explanation of the fact may be, the important point is to show that it exists, and that there is a difference in light, so far as its *quality* is concerned, at the same hours of the day, at the north and in the heart of Europe. But this is not all. After having examined this question of quality, we have to look next at the question of *quantity*. The days which are useful to vegetation at the north are longer than with us—what now is the influence of the duration of light on the chemical phenomenon which serves us as a means of measurement? Is the effect thus produced proportionate to the length of exposure to the sun? Does it increase more or less slowly? Such are the first questions which we have to consider.

I believe they are new, because up to this day, both as regards meteorologic instruments and in theoretical speculations, it has always been held that the effect



of illumination was, everything else being equal, proportional to its duration. We shall see that this is not so, and that the effect increases much more rapidly than the increase of time, so that all the notions which we entertain on this subject stand in need of revision.

#### INFLUENCE OF THE DURATION OF ILLUMINATION.

So far we have taken as a measure of the total actinic effect during the period of exposure, the sum total of the oxalic acid consumed. The conclusions which we have thus reached, subsist, whatever may be the law which connects the combustion with the actinic effect; it has been enough for us to expose, during the same length of time, solutions equally sensitive, in two different places and to proceed always by comparison.

But the law of the increase of actinic effect, with the time of insolation, does not the less merit attentive investigation. To begin, let us ask first, if the total effect of combustion, observed at the close of a day, upon a solution of oxalic acid exposed to the sun, represents the sum of the divers actinic effects produced at the different hours.

One way to answer the question is to expose in the morning, side by side, two vessels holding the same quantity of the same solution. One is to be examined at the end of the day, and this will give us the sum total of the effect. The other is to be examined at the end of an hour, and then to be replaced by another vessel like the first, but containing new liquid, to be likewise studied after another hour's exposure. In like manner, we shall renew the study at successive hours. If the actinic effects accumulate, without loss or encroachments in the liquid of the vessel which has been exposed to the sun all day long, the quantity of acid which we shall find has disappeared must equal the sum of the quantities of acid which have vanished in the vessels that were exposed for an hour each.

The experiment, repeated again and again, shows that it is never so. The sum of the quantities of acid burnt in the vessels which have been exposed each one hour only, is always insignificant in comparison with that consumed in a vessel which has spent the whole day in the sunlight. The difference varies from one day to another, and increases with the intensity of combustion. It decreases slightly if we carry the exposure of the successive vessels to two hours, and still more if we extend it to three or four hours, as should be expected. But, even if we divide a day of ten hours into two equal periods, one from 7 A.M. to noon, and the other from noon to 5 P.M., the sum total of acid consumed in the two vessels that correspond to the two periods of exposure sometimes does not exceed half the acid consumed in the vessel which spent ten hours in the sun. The combustion, therefore, does

not begin as soon as the vessel is exposed to the light. There is a "lost time" at the beginning; two hours, three hours, are necessary for the solution to put itself in action. During this whole time the work is wholly interior and betrays itself by no diminution of the acidimetric titre.

This "dead time" at the beginning of the reaction should not surprise us. When we study the different reactions of chemistry, from this point of view, we become aware that there are few which begin immediately upon realization of the exterior conditions of production, even in cases when the energy which comes into play is altogether internal as regards the mixture. The formation of a precipitate of barium sulphate is not instantaneous; that of calcium sulphate, or of calcium tartrate, is still less so; a mixture of formate and of permanganate of potassium remains apparently inert for some seconds, after which begins an abundant, and, to some extent, an explosive liberation of carbonic acid, proceeding from the combustion of formic acid.

Here the heat produced by the reaction intervenes to increase its activity. We can say the same of the phenomenon which Bunsen and Roscoe discovered and investigated under the name of "photochemical induction," in the combination, in the sunlight, of chlorine and hydrogen. This reaction also requires a certain time to commence, but it accelerates afterwards, because it is exothermic. The same remark applies to the reduction of chloride or bromide of silver in the presence of an organic substance, which also shows a "dead time" at the beginning, and becomes more energetic afterwards. The same remark applies, moreover, to almost all photographic operations, whether we wish to obtain luminous impressions, to develop images, or to produce positive prints.

If we observe a "dead time" when the forces are internal and accelerating, it is not surprising that we should find a like one also in the solar combustion of oxalic acid, where the impulse is to come from without and where the reaction is so feebly exothermic. But this verification presents here an interest which it has not elsewhere, for we connect it intimately with the phenomena of sensibilization, which we discovered previously in the solutions of oxalic acid. In both cases a *molecular preparation* is evidently involved, the mechanism of which is still unknown to us, but which results in placing the molecule upon a kind of inclined plane, down which it may be made to roll by the slightest impulse. As a confirmation of this idea, I have ascertained that in fact the "dead time" at the beginning is less protracted with solutions which have been made sensitive, than with new solutions, so that if the latter do not undergo in the light of the sun, as we have seen before, the same degree of combustion as the others, it is partly because the "dead time" at the beginning is shorter. But I say "partly" because there is still another



phenomenon. We shall see that combustion, once begun, does not go on with regular and equal steps, but is made to proceed faster and faster. In other words, everything goes on as if the sensitiveness were increasing during the oxidation. To put it still differently, the quantity of burnt oxalic acid, which amounts to little or next to nothing during the first moments of the exposure to the sun, starts out and increases, from that instant, quicker than time, so that there is no proportion between the length of the insolation and its consuming effects.

In order to take account of effect of insolation upon an oxalic solution, let us slightly modify the conditions of an experiment which I have just described. Let us expose in the morning a dozen similar vessels to the sun, and let us withdraw every other hour two of them, which will give us the sum total of combustion up to that moment. It will be easy by this means to ascertain the progress of combustion during the whole day. The following experiment I cite, not as being the most complete of those which I have made, but because it was performed with a solution of the same sensitiveness as that used in other experiments which I shall quote presently, so that all of them are comparable.

*Exp.*—On September 6, 1888, at 8.30 A.M., on Mont Dore, I exposed to the sun four vessels, which I withdrew at various intervals, and in which I measured the proportion of oxalic acid burnt.

		Solar Combustion %
After 2 hours		0 %
" 4 "		3 %
" 8 "		10 %
" 10 "		12 %

We see at the start the "dead time" of the beginning. We see, moreover, that from the fourth to the eighth hour, that is to say from 12.30 P.M. to 4.30 P.M., the combustion was twice as rapid as from 10.30 A.M. to noon, in spite of the gradual descent of the sun towards the horizon. During the last two hours, and notwithstanding the obliquity of the solar rays, which is already great at this time of the year, the combustion was still two thirds of what it had been between 10 A.M. and noon.

It is always the same, whether the total combustion be feeble, as it was at Mont Dore, or active, as I have at times found it in Paris. From the sum total of my results I think I may conclude that the progress of solar combustion does not remain constant during the whole of the day, and that instead of increasing towards noon, in order to decrease afterwards in proportion as the sun approaches the horizon, it, on the contrary, experiences a progressive acceleration which does not cease till the sun is near its setting.



Everything, then, goes on as if the sensitiveness which, as we have seen, a solution of oxalic acid attains if left to itself in darkness, were not by any means a maximum sensitiveness, and might be greatly increased in the light. I have in fact ascertained—and we shall presently see an example of it—that a recent solution of oxalic acid may be made very sensitive by a few hours' exposure to the sun, and so be brought up to the level of an old solution, or even beyond it. But then an unforeseen consequence appears: the sensitiveness acquired in darkness is persistent—might it not be perhaps the same with sensitiveness acquired in the light, so that the accelerating effect of a fine day might spread, as it were, in its totality, or at least in part, over the following day?

The following experiment proves in fact that a solution left for a whole day in the sun, and which has not been entirely oxidized, will retain for the next day a greater sensitiveness than another part of the same solution which was not previously insolated.

*Exp.*—There were exposed every day to the light four identical vessels, two of which were carefully examined at the close of each day, while the other two were left in reserve for the day following; on this day they were again exposed to the sun at the same time with the four new vessels of a second experiment. The total of combustion in the vessels which were exposed for two days was then compared with the sum of combustions in the vessels which were each exposed only one day. Some of the results which I thus obtained are as follows:

The day of September 2d, combustion	10 %	} 34 % in all.
" " " " 3d, "	24 %	
Both days together,	68 %.	

The amount of combustion has therefore doubled. Here are the results of another experiment:

The day of September 4th, combustion	12 %	} 23 % in all.
" " " " 5th, "	11 %	
Both days together,	38 %.	

The difference points in the same direction as the preceding experiment; only it is not quite as great, because the two days, September 4th and 5th, were both quite indifferent days (page 28), whilst the day of September 3d in the first experiment was very fine.

To sum up, we see that the insolated vessel of the first experiment underwent on the second day a combustion of  $68 - 10 = 58$  per cent, while a new vessel suffered only an oxidization of 24 per cent. As to the second experiment, the cor-

responding figures are 26 per cent and 11 per cent; this shows that not only does the sensitiveness of the oxalic solution increase in consequence of insolation, but the gain continues during the night. Some experiments of the same kind, which I will not now explain in detail, prove that this excitation of sensitiveness by means of insolation, endures even to the second day after, in a solution which is kept in the dark after having been exposed for a day to the sun. It is only after three days, therefore, that traces of sensitiveness are no longer discernible. By that time, the insulated solution has nearly returned to the degree of sensitiveness which the mother liquid possessed, which seems thus to correspond to a kind of equilibrium. It is in fact remarkable that the different sensitive liquids which I have used in my long experiments and which were prepared at very different times, with the single precaution that they were not to be used before the lapse of several months, had all of them, at the moment when I used them, very nearly the same sensitiveness. It was on September 8th and 9th that I had to change my solutions at Mont Dore, and I availed myself of the fact that these two days were but indifferently fair, to interrupt my series and to compare again and again the old liquid with the new. The titre was always the same for both. Mr. Elfving compared likewise two solutions which I had sent him a year apart, and found in four days of experimenting the following corresponding figures for the old and the new:

	Old Solution.	New Solution.
1st day, combustion,	58 %	56 %
2d " "	52 %	51 %
3d " "	63 %	60 %
4th " "	42 %	35 %

The old solution was a little more sensitive, which is the usual rule. But the difference was trifling, and thus our former statement was confirmed (page 9). The oxalic solution, kept in diffused light, reaches a fairly constant sensitiveness in a few weeks, but this maximum, although stable, is not a *maximum maximorum*. It may be temporarily exalted in the sun, continue if the illumination continues, and return to its original level after some days of darkness.

In order to make this conclusion really valuable, we have to overcome one last objection. Might it not happen that the increase of combustion discovered on the second day in a vessel which had been insulated on the day before, might mean simply the suppression of the "dead time" at the beginning? Starting earlier, the combustion might better utilize the good hours of the day, and thus be enabled to go farther. A priori, the intervention of this cause does not seem to explain sufficiently the great difference observed. But it is safer to consult experience. It will be

sufficient to cut the two days of observation by an intermediate examination and to discover the intensity of combustion during each one of the intervals. Thus it can easily be seen whether the vessels change at the same rate, after having recovered their "dead time," or whether the vessel which had been insolated on the previous day, still progresses more rapidly than its neighbor?

*Exp.*—On September 12, 1888, a fine day with a warm sun, and a few cumuli, there were exposed to the sun 4 vessels, Nos. 1, 2, 3, and 4.

Vessel 1 was examined

after 5 hours; combustion 10 per cent.

Vessel 2

after 9 hours; combustion 18 per cent.

Vessels 3 and 4 were put aside and exposed anew on the next day with two new vessels, Nos. 3' and 4'. This day, the 13th, was very fine, with a few cirri in the morning. It changed a little for the worse towards evening.

Vessel 3' was examined

after 5 hours; combustion 13 per cent.

Vessel 3

after 5 hours; combustion 44 per cent.

The difference is considerable and is certainly in part at least due to the suppression of the "dead time" in the vessel which was insolated on the day before. But this again is not all, for during the second half of the day, the insolated vessel kept up a much more rapid progress than the other, as the following figures clearly show:

Vessel 4' examined after 9 hours; combustion 25 per cent.

Vessel 4 examined after 9 hours; combustion 62 per cent.

The acceleration in the solution which had been insolated on the day before, thus continued throughout the day, and while in the second half of the second day, the new liquid only showed an increase of  $25 - 13 = 12$  per cent in its combustion, the liquid insolated the day before rose from 44 to 62, undergoing thus an increase of 18 per cent.

It will also be noticed that in the morning this same liquid had increased from  $44 - 18 = 26$  per cent, while the new liquid experienced a combustion of only 13 per cent. Here has come in the double effect of suppression of "dead time" and that of the acceleration. The two solutions became a little more nearly equal towards evening, but the insolated solution continued its quicker progress.

Thus there can be no doubt that the insolation during the previous day continued its effects over the next day and the day after that. But this is not all. One fact, no less curious than the preceding, is that the sensitiveness due to the



action of light, enables the solution to undergo in diffused light a combustion which is out of question as long as it has only its normal degree of sensibility, that is, the degree obtained by keeping for some weeks in a diffused light.

*Exp.*—In August, 1889, at Noalhac near Aurillac (Cantal), at an altitude of about 700 metres, I prepared a solution of oxalic acid, part of which was left in a flask, exposed to a very feeble light, while another portion was exposed to the sun in a stoppered bottle. This was in order to see if the process of rendering the solution sensitive was necessarily accompanied by a process of combustion, or whether it could be accomplished without the latter. The experiment showed that the two effects are independent of each other. The solution contained in the closed flask had at its disposal only a very small quantity of dissolved oxygen, which it consumed, moreover, with but a slight diminution of its titre, but in one day of insolation it reached an intense sensitiveness, which was maintained for several days at the same rate by preserving it in a diffused light.

On August 30th, I exposed to the sun two vessels containing some of this insolated solution, and at the same time, two vessels of the same non-insolated preparation. The figures for solar combustion are:

18 and 19 per cent for the non-insolated, and  
92 and 92 “ “ “ insolated liquid.

The day was a very fine one; the flask with the insolated solution remained in the sun, but closed. On August 31st, two groups of two vessels each were prepared, one containing non-insolated, the other insolated solution. One of these groups was exposed to the sun, the other on a window-ledge, facing the north, where it received no light but that coming from the sky and dimmed somewhat by a slight dry fog. The figures found after 8 hours' exposure, for combustion in the sun and in diffused light, were the following:

Insolated	liquid lost 63 per cent in the sun.
Non-insolated	“ “ 24 “ “ “
Insolated	“ “ 19 “ in diffused light.
Non-insolated	“ “ 6 “ “ “

The next day, September 1st, during a dark, threatening day, the same arrangement gave the following results:

Insolated	liquid lost 50 per cent in the sun.
Non-insolated	“ “ 13 “ “ “
Insolated	“ “ 6 “ in diffused light.
Non-insolated	“ “ 3 “ “ “

Thus a previous insolation increases the rapidity of combustion, not only in direct light but also in diffused light. The experiment of September 2d shows, however, when compared with that of the day before, that this diffused light must have a certain intensity to make its effect measurable after some hours. But, viewed by itself, this experiment shows that even a dark day still has an accelerating effect upon a solution which has not seen the sky since the second day before.

The insulated liquid was from that moment kept in diffused light at the back of a room with but one window, facing the north. It was found that after a few days it had not sensibly changed in titre, but variation began to show itself at the end of a month. We thus see how here also, in spite of good conditions of preservation, the phenomena of slow combustion appear which have been observed since Wettstein in solutions of oxalic acid. Not insulated, this solution preserved in the same manner had remained much more stable, which shows that it is necessary to avoid exposure to light, even temporarily, or even in a carefully closed flask, of solutions of oxalic acid which are intended for processes of titration. The luminous impression, once received, persists and makes them much less stable—it continues, as we shall presently see, even after the liquid solution has been placed in darkness.

*Exp.*—Another experiment was begun identical with those that have just been described except that the flask which contained the insulated solution, sheltered from the air, was kept for three nights and two days in a cupboard of the laboratory before being distributed into vessels on September 6th. Unfortunately the day of the 6th was disturbed by cirri and cloudlets.

The insulated liquid lost 20 per cent in the sun,  
Non-insolated liquid lost 7 per cent in the sun.  
The oxidation in diffused light was insignificant.

The proportion of oxalic acid burnt in sunlight is, therefore, still, after 60 hours of obscurity, three times greater in the insulated solution than in the other. But the sensitiveness decreases afterwards and the difference soon ceases to be measurable after a day's exposure. We here meet again with that retrogradation which we have pointed out earlier, and which brings us back to normal sensitiveness.

I add, in order to close the subject, that this solar impression, which disappears slowly, is on the other hand produced very rapidly, and that, when investigating comparatively, with respect to the combustion which they undergo, the solution which I had exposed to the sun, in three flasks, and 1, 2 and 3 days, respectively, I have not been able to show that there was any essential difference between them!

I have left aside, in all which precedes, the question of the mechanism connected with both sensitization and combustion. The former goes on when the solution is sheltered from the air, and can take place only by a new arrangement of molecules. Combustion, on the other hand, takes place in contact with the air and possibly with the formation of ozone or of hydrogen peroxide. That is a question which must be investigated by itself. I purpose here only to put in evidence, as regards the constitution of the atmosphere, some properties and a variability of effects, not hitherto observed.

#### METEOREOLOGICAL, HYGIENIC, AND AGRICULTURAL EFFECTS.

If we now return, with the results which we have obtained, to our investigation of the causes which provoke the rapid development of vegetation in the extreme northern regions, we see that those regions are superior to ours in a twofold aspect.

1. That cause which depends on the constitution of their atmosphere consists in this, that the absorption of the chemical radiations of solar light is there less great than with us. The actinic power at the level of the soil exceeds that which we have observed around ourselves at different hours of the day, and that in spite of a lower sun and a greater thickness of atmosphere, which the rays must traverse. These differences are due mainly to the fact that vegetation in the north sends into the air fewer oxidizable substances to form a screen.

2. The other point of superiority connected with the geographical situation consists in this, that in the extreme north, during the period of vegetation, the days are longer than in our temperate zones, and that the actinic power, at least so far as it may be measured by a solution of oxalic acid, increases more rapidly than the length of the day, and this out of all proportion. After a period of preparation, combustion begins, then accelerates so rapidly as to make up for time lost at the beginning, and finally, towards evening, reaches unusually high figures, such as are unknown to our regions. It is in this way that combustion has risen to

37 and 89 per cent on September 2 and 4, 1887,

79 and 80 per cent on September 9 and 11, 1888,

75 per cent on Aug. 27, 1888 at Helsingfors.

and this at a time when the highest figures, relatively to the same periods and with the same solutions, did not exceed 50 per cent and were even sometimes much lower in our country. In order to reach figures equal to those obtained in the Gulf of Finland, it was necessary for me in France to accumulate upon my vessels the



radiation of two consecutive days. It may thus be said that, speaking generally, a day at the north is worth two of ours as regards actinic power.

Not only does the actinic effect of a fine day increase more rapidly than the length of the day itself, but it may actually spread itself over the next day, and the day after that, and thus make up, in some degree, for the absence of the sun. In like manner, a fine morning may render combustion more rapid even though the evening be dark and stormy. It is enough that the liquid shall have been made sensitive; and as this sensitization is the more rapid as the actinic intensity is greater, the atmospheric condition of northern countries favors them in this respect beyond us, and a new superiority is thus attained through the superposition and mutual emphasis of the other two causes.

Finally, the sensitiveness produced by a fine day continues for several days. A number of bad days, following each other, is consequently not a period of inertness and loss; it draws upon the store which was collected during fine weather. On the other hand, we have seen that the sensitiveness which was acquired in the sun, did not increase without limit, and that it reached quite rapidly a maximum beyond which it did not go. A succession of fine days, therefore does not develop actinic phenomena to an extreme. We here meet once more with the system of balancing which weakens great effects, increases small ones, and which has been pointed out with regard to so many other manifestations of the forces of nature.

Summing up the matter, then, it would seem that we have hitherto missed our way, in considering the chemical action of solar light as independent of locality and proportionate to time of isolation, or as furnished or measured by meteorological instruments. The first of these notions was purely instinctive and was suggested especially by the uniformity which was ascertained to exist at different points of the globe in so many other grand meteorological phenomena (such as the composition of the air, the average barometric elevation, the mean distribution of nebulosity, etc.) Instead of such a uniformity, we find, on the contrary, actinic climates, limited in point of surface, for they betray the local influence of the surface of the soil—limited also in point of duration, for they are due to two kinds of clouds which are subject, like the others, to the influences of place and season.

Misjudging thus local influences, only the first cause has been thought of, and all efforts had been directed towards measuring the duration of insolation. On this point, I think I have shown that the wrong way had been taken. The actinic force of a day is not the same for the same day, in different parts of the globe, and its effect increases more rapidly than its length; such is the principal lesson of this Memoir.

One step farther might be taken. We have just ascertained that in the solution of oxalic acid there takes place a kind of storing up of light, which shows itself in an increase of sensitiveness as regards phenomena of oxidation. Might not the oxygen which is present in the solution, or even that which is constantly dissolved there and transformed, might it not itself be rendered sensitive, and so be endowed with an oxidizing power which it could afterwards use in diffused light?

I have found nothing, while searching in this direction, with oxalic acid; this reagent, quite sensitive enough for the study of powerful actions, is not sensitive enough for such weak actions as that which I have just suggested. But I have been more successful with oxidizable substances of sharper reactions, so that the very smallest variations became measurable. This is the case with diastases; an almost infinitesimal quantity will produce very apparent effects, and it is, therefore, easy to trace their disappearance by oxidation in the liquids which contained them. With rennet especially the very smallest variations in quantity can be appreciated from corresponding variations in the time of coagulation of equal quantities of milk, so that this diastase is very convenient for study. By such means I found that it oxidized and disappeared in water which had been previously exposed to the sun, while it remained, if not quite intact, at least nearly so in the same water as freshly drawn from the hydrant. I also found that a glass flask exposed to the sun stored up on its walls enough chemical radiations to accelerate afterwards the oxidation of a solution of rennet, which was allowed to stand in the shade.

All these facts, upon which I do not insist because they go beyond the limits of this work, enlarge the field of those phenomena to which they apply. If insulated liquids and solids, may in certain cases, like our solutions of oxalic acid, acquire properties which they had not before, the phenomena of solar combustion may well extend below the surface, which has been directly illuminated, and assume in the general economy of the world an importance, no doubt as yet inferior to that of microscopic organisms, but certainly no longer to be neglected, as it has been heretofore.

Since I have entertained the views which I have developed in the preceding pages, I have investigated especially their agricultural and hygienic consequences.

As far as hygiene is concerned, I have shown more clearly I think than Messrs. Downes and Blunt that solar light kills the germs of microbes suspended in the air. I have proved, moreover, that this destruction was preceded by a veritable *attenuation*.<sup>1</sup>

<sup>1</sup> *Annales de Chimie et de Physique*, 6th ser., vol. v., 1885.



I have ascertained since that this destruction and this attenuation go on in the superficial layers of the soil and even down to some depth. If, in the many attempts to count the microbes of the soil, it has so often been found that the number is less near the surface than at the depth of a few centimetres, we must attribute this result much rather to solar combustion than to desiccation. The sanitary action of oxygen, which is pursued and completed in the atmosphere, begins therefore, thanks to light, at the surface of the soil, and the healthiest countries are those in which the actinic power of the sun is greatest.

By a curious mechanism, which I have tried to make generally known, the solar action which neutralizes the microbes which it encounters, can act like them, and take their place. I have in fact shown in an extensive work<sup>1</sup> that the changes which carbohydrates undergo upon exposure to sunlight are exactly like those which they undergo under the action of ferments. Starting from the same point, these two modes of transformation, apparently so different, resemble each other not only in their variety and marvellous flexibility of conduct, but still more in their intermediary and final products.

Thus, invert sugar in alkaline solution, oxidized in sunlight, gives intermediary products which are colloidal and identical with humic acids, except that they are not nitrogenous. These black acids are afterwards consumed by light, exactly as we see in the bleaching of the black soil which the spade or the plow has turned up.

The extreme terms of the transformation of this sugar or of its humic derivatives are as numerous and as varied in solar combustion as when produced by the action of ferments. Thus, by contact with potash, or with soda, we obtain alcohol through an interior combustion which is identical with that produced during alcoholic fermentation. On the other hand, in the presence of baryta, no alcohol, but lactic acid is produced. In this there is analogy, not with alcoholic fermentation, but with lactic fermentation, and this analogy is all the closer since—as is recognized—there may be several lactic acids of different rotary power, which may be produced as well by the action of light as by that of fermentation.

This solar, lactic fermentation is accompanied by the production of acetic acid, as in the case of microbial fermentation. In other cases, butyric acid is formed, formic acid, oxalic acid—in short, all the ordinary residues of the ferment action. Finally, carbonic acid represents in all cases the extreme term of the change of organic matter into gas.

The luminous action, varying in quantity according to place and season, as the

<sup>1</sup> *Annales de l'Institut Agronomique*, vol. x., 1886.



different chapters of this Memoir have shown, may therefore differ in the quality of the effects which it produces. All these facts lend to the study of chemical radiation, an importance of the highest rank, and I shall consider myself very happy, if the first results contained in this paper shall lead men of science to new researches.

### SUMMARY.

1. The oxidation of oxalic acid in a weak solution takes place mainly, and almost exclusively, under the influence of the chemical rays of solar light; it can, therefore, be used as an actinometric measure.

2. It depends on the concentration of the liquid, which for the best results should not exceed about three grammes per litre.

3. With an equal volume of solution, combustion decreases as depth increases; there is an absorption of chemical rays, although the liquid is and remains very transparent.

4. For equal depths of liquid, combustion is proportional to the surface, and consequently also to the volume.

5. It depends on the age of the solution, that is to say, of the time which has elapsed since preparation. As it grows older, an oxalic solution becomes more sensitive, and attains a certain maximum which is quite stable and quite regular. It is well to wait till this state of sensitiveness has been produced.

6. The daily combustion, such as is measured with sterilized liquids, varies from one day to another much more than any other meteorological phenomenon, and while subject to the influence of what we call "fine weather" and "overcast weather," it manifests very clearly other influences which are less visible.

7. It shows also the influence of the seasons, and manifestly exhibits a maximum in spring.

8. It is but feebly subject to the influence of altitude.

9. On the other hand, it betrays so strongly the presence of divers oxidizable essences or substances in the air, that we must consider local and daily variations as due to the presence in the atmosphere of actinic clouds, which are discoverable only by the reduction and absorption which they produce in the chemical radiations of sunlight.

10. The atmosphere of extreme northern regions is less absorbent than that of our temperate zones, and, consequently, at the same hours of the day, actinic radiation is more powerful, at the level of the soil, in the north than at the centre of Europe.

11. Northern countries add to this cause of superiority, which they owe to the constitution of their atmosphere, another, which is due to their geographical position, namely : that the actinic effect of the sun increases more rapidly than the duration of its presence above the horizon. The very long days of the north, during the period of vegetation, are, therefore, in their actinic effect, more active than an equal number of days in our temperate regions, and we can thus explain the particularly intense rate of the progress which vegetation makes in the vicinity of the polar circle.

12. This increase of sensitiveness which oxalic acid experiences in the sun, does not cease when the light begins to fade, and may continue several days. Hence follows a conclusion which may also be applied to our temperate regions : this is, that the actinic effect of a number of fine days in succession increases more rapidly than its duration, and also, that the effect of a fine morning is not lost by a dark and cloudy evening.

13. We must, therefore, give up the hope of finding, in the duration of a day or of solar action, a measure of its effects, and meteorological instruments, which accept such a proportionality, are to be rejected.

14. The importance of these actinic phenomena in the general economy of the world is great enough to make it necessary that we should approach the investigation by appropriate means.

## SUPPLEMENT.

### OBSERVATIONS MADE IN 1894 IN FRANCE AND ALGIERS.

Since sending my Memoir on Atmospheric Actinometry, I have been enabled by the courtesy of M. Gessard, Chief Pharmacist of the Military Hospital at Setif (Algeria), to make a number of combined observations in a temperate region and in a hot climate. It was interesting to discover whether we would meet here with the same differences as between the observations made in France and in Finland, that is to say, if for equal lengths of insolation the chemical activity of the solar rays would continue to diminish in proportion as we approach the equator, and as their calorific power increases.

For such a comparison the choice of the stations was of some importance. Setif is situated about eleven hundred metres above the level of the sea, on a buttress of the southern slope of the high mountains of the sea-coast, the chain of the Babers or the Bibans. Towards the south it overlooks from a height of two or three hundred metres an immense plain, which in its turn is bordered at a distance of 35 or 40 kilometres (22 to 25 miles) by a chain of not very high mountains, which cuts it off from another more extensive plain, the basin of the Hodna. Beyond this, separated again by an insignificant mountainous elevation, lies the Sahara and the desert climate, which makes its influence felt as far as the plain of Setif. This vast heating-centre south of the city frequently procures for the latter, towards evening, a fresh current of air from the north, and in ordinary times it stands on the boundary line where two contrary influences enter into direct conflict, the wind blowing from the coast and the high summits, and the burning wind from the desert. Thus Setif enjoys a relative freshness on certain days when the plain at its feet is given up to the full ardor of the sirocco, and when it even may happen that the cloud of dust, propelled by this wind, stopping at a distance of 15 or 20 kilos. from the town, screens the neighboring mountains at the very time when the atmosphere remains quite clear about Setif and the immediate surroundings.

The station which I have chosen in France for my comparative observations is also situated on the side of a slope, overlooking the plain of Vic-sur-Cère (Cantal), and 750 metres above the level of the sea. I might have gone higher, but I have



already shown that the difference in altitude is of little importance. At all events it acted in the opposite sense to the phenomenon which I sought to verify. As a compensation, the climate of this station at Olmet is a temperate climate. The place lies on the line where the culture of the vine ceases and is in every respect equal to that of Fan, in the valley of Marmanhac, and of Noalhac, in the valley of Aurillac, where I had made my first observations.

Furthermore, the procedure of M. Gessard and myself was the same; we exposed, from 8 o'clock A.M. till 5 o'clock P.M., vessels containing the same solution, only, on account of the high temperature of Setif during the summer and of the evaporation caused by it, we had to pour into the vessels 20 cubic centimetres of oxalic acid, instead of 10, and place them, not upon wooden or stone supports, but upon the water of a great crystallizing pan. I need not say that at Olmet I followed the same practice. It is well known that the degree of solar combustion depends on the depth of the liquid, and this is the reason why the present series of experiments is not directly comparable with the preceding series. But it is sufficient for us that the experiments made in France and in Algeria should be comparable between themselves.

This being granted, I subjoin the results obtained by M. Gessard:

Date.	Solar Combustion.	Remarks.
July 2	12	
" 3	14	
" 4	12	Clouds.
" 5	13	Stormy weather. Sky covered at 3 o'clock.
" 6	16	Fewer clouds than on preceding days.
" 7	17	Clear weather. No dust.
" 8	62 <sup>1</sup>	Cloudy at times. (See note below.)
" 9	18	Tempest. Dust. Sun obscured after noon.
" 11	40	Cloudy weather.
" 12	32	Fine weather.
" 13	22	Overcast weather; a few drops of rain.
" 15	25	Clouds, storms, dust repeatedly.
" 17	22	Fine weather. Hail-storm at 7 P.M.
" 18	22	The Southern mountain cannot be seen at 5 P.M.
" 19	24	No clouds. Very clear horizon.
" 20	22	Overcast from 2 to 4.30 P.M. Dust.
" 21	23	Overcast after noon. Downpour at 3 P.M.

In the following experiments, made in August and September, M. Gessard ascertained the direction of the wind and the temperature, as read on a thermometer

<sup>1</sup> One of the vessels was found to be submerged on this day, so that the result could not be correctly ascertained. The weather on this day did not essentially differ from that of the preceding and of the following days.

hanging in a northern exposure, against the wall of a house with lofty arcades, and consequently under a gallery formed by them. In the statements concerning the winds, the frequent violent changes of which we spoke at the beginning will be noticed.

Date.	Solar Combustion.	Remarks.	Temperature.			Winds (by the vane) at			
			Max.	Min.	Aver.	8.30	11	3	5
August 18	6 %	Clouds at noon. Overcast sky at 4. Rain at 4.30.	34.	15.2	31.5	W	W	N	SW
" 19	3 %	Quite a fine day. g. g. clouds at 2 o'clock.	30.2	14.	27.5	E	S	NNE	NNE
" 20	3 %	Sun rather overcast in the morning. Rain after 2 o'clock.	27.9	12.5	25.	SE	N	N	NE
" 21	3 %	Sun all day long and sky clear.	28.6	15.8	25.5	W	W	NW	N
" 22	8 %	The same.	31.1	17.5	28.5	W	W	N	NE
" 23	8 %	" "	32.6	18.6	30.	SW	W	S	SW
" 24	11 %	" "	33.2	17.2	31.5	SW	SW	S	SW
" 25	6 %	" "	31.4	17.	19.	SW	W	S	SW
" 26	8 %	Clouds in the afternoon.	32.6	17.8	29.5	SW	SW	S	SW
" 27	9 %	Clouds afternoon. Sun shines at 3 P.M.	31.8	16.2	29.5	W	W	NE	NE
" 28	8 %	Sun all day long.	31.3	16.3	28.5	W	SW	NNW	NNE
" 29	7 %	Same.	31.1	16.3	29.	SW	SW	SW	NE
" 30	7 %	"	31.6	18.6	30.	SW	SW	SW	NE
" 31	7 %	"	32.2	19.0	29.5	SE	SE	SE	SSE
September 8	9 %	Overcast. Much dust.	25.4	17.	29.5	E	S	SSE	S
" 19	10 %	Sun all day long.	29.	15.6	29.5	SE	S	SSE	S
" 22	11 %	A few clouds all day long.	29.	14.8	27.	W	NW	W	N
" 23	11 %	Sun all day.	29.4	17.7	26.5	W	NW	W	NW
" 24	11 %	Sun rather obscured, afternoon. Dust.	30.9	17.1	28.	SW	SW	SW	SW
" 25	14 %	Overcast sun all day. Dust.	30.8	17.	28.	S	SW	SW	S

When M. Gessard sent me the reports for August and September, he added what follows: "They seem to me to confirm the former. The combustions have not been more intense than in July. I notice, on the other hand, that during overcast weather they have not been inferior to those observed on days of full sunshine. The second fact which strikes me is this: that the quality of combustion should be maintained with such constancy in the two vessels under all circumstances. Thus, on the 11th September (I did not report it in the table), with a south wind blowing all day, raising the maximum temperature to 32° 4', the contents of both vessels having evaporated, although they had been allowed to float upon the water, I had the curiosity to re-dissolve the residual acid and to test it: I found 8.4 c. c.

and 8.6 c. c., which corresponds to combustions of 36 per cent and 35 per cent, respectively.

It will be seen from all that has been said so far, that the daily actinometric combustions are quite as irregular in Algeria as in France, and have only a very remote connection with the external aspect of the sky and the clearness of the horizon. I have not been so fortunate as M. Gessard so far as the weather was concerned. The greater part of the month of August in the Cantal and a part of the month of September we had cloudy or rainy periods, and during this time observations were impossible. Availing myself of the fact that the method of comparison which we employed consists of a comparison of the finest days at the two stations, I shall here report only the figures noted down at Olmet during the rare times of fine weather.

Date.	Solar Combustion.	Remarks.
August 27	29 %	Fine day from beginning to end.
" 28	42 %	Fine weather, but rather heavy and stormy.
" 29	41 %	A fine day, although rather foggy.
" 30	35 %	A fine day, rather stormy. Some clouds.
" 31	41 %	A fine day. Some mist towards 4 o'clock in the west.
September 2	27 %	Clear in the morning ; rather foggy in the evening.
" 17	40 %	Quite a fine day in spite of g. g. clouds. N. E. wind.
" 18	34 %	Cumulus covering $\frac{1}{4}$ of sky, all day long.
" 19	52 %	Less cumulus than the day before. Finer day.
" 20	60 %	A very fine day.
" 21	52 %	Rather stormy day.
" 27	28 %	A fine day ; few clouds.
" 29	23 %	An indifferent day.

Comparing this list with M. Gessard's we notice in the first place a coincidence between the days of August 27th, 28th, 29th, 30th, and 31st, which were either fine or very fine at Olmet and at Setif. Now if we compare the results of the daily combustion from 8 A.M. till 5 P.M. we find, respectively :

9, 8, 7, 7, and 7 for Setif ;  
and 29, 42, 41, 35, and 31 for Olmet.

If we in like manner compare, leaving out the dates, the combustion on the finest days in France and in Algeria, we find again that it decreases with the latitude. This is the same conclusion to which we were led, when we compared the observations made in France and in Norway, and between the limits of the two stations at Helsingfors (lat.  $60^{\circ} 19'$ ), and at Setif (lat.  $36^{\circ} 11'$ ). This shows that the



actinic effect of twilight during equal times of insolation goes on diminishing in proportion as we approach the equator and as the mean temperature rises.

It would be interesting to ascertain whether this law continues into the tropical regions. This is probable, but as yet is only a matter of conjecture. I have taken the proper steps to begin observations on this subject early next spring.

Whatever this extension may lead to, it is none the less unexpected to find that in our temperate regions, the most densely populated of the globe, the actinic effect of the sun is, so to speak, in the opposite direction to its calorific influence. The operation of this law on the flora cannot be doubted. But the future alone can tell us how this is brought about.



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